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Antiferromagnetic Spin Fluctuations and the Heavy Fermion System UPt_3

A. I. Goldman and G. Shirane
Brookhaven National Laboratory
Upton, New York 11973

G. Aeppli
AT&T Bell Laboratories
Murray Hill, New Jersey 07974

E. Bucher and J. Hufnagl
University of Konstanz, D7750 Konstanz
Federal Republic of Germany

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ABSTRACT

We review the results of inelastic polarized and unpolarized magnetic neutron scattering measurements on the heavy fermion superconductor UPt_3. Below T_χ = 18K we find evidence for antiferromagnetic spin fluctuations with a modulation vector along the c-axis of the hexagonal lattice. This contradicts the analogy often made between UPt_3 and liquid ^3He, and may have important consequences for the pairing mechanism responsible for superconductivity in this system.

The heavy fermion system (HFS) UPt_3 has been the subject of intense experimental and theoretical efforts.[1] Like the other HFS which become superconductors at low temperature, many of the superconducting properties of UPt_3 are non-BCS like. This has led to the speculation[2] that some exotic pairing mechanism is responsible for the superconductivity in these materials. Of equal interest is the behavior of many of the low temperature normal state properties of UPt_3. In particular, there is a maximum in the bulk magnetic susceptibility measured[3] in the ab plane of the hexagonal lattice at $T_\chi = 18K$. Below this temperature, in addition to the large linear electronic contribution to the specific heat $\gamma T (\gamma = .45J/mole\text{-}K^2)$, there is also a contribution that may be described as $T^3 \ln(T/T_0)$, which has been used as evidence for the onset of ferromagnetic spin fluctuations in this low temperature regime, as for liquid ^3He. Therefore, macroscopic measurements indicate that UPt_3 changes in some fundamental way below $T_\chi = 18K$.

We review the results of neutron scattering measurements[4,5] on single crystal specimens of UPt_3 to investigate the energy and wavevector dependence of the paramagnetic response at low temperatures. For $T \leq T_\chi$ we have found strong evidence for antiferromagnetic (AF) correlations between the U-moments which disappear above, at most, 30K.

Two cylindrical single crystals cut from the same boule, with c* roughly parallel to the cylinder axis, were mounted over each other and aligned with their c* $(00Q_Z)$ axis in the scattering plane. Unpolarized beam measurements[4] were made on a triple axis spectrometer using a pyrolytic graphite (PG) (002) monochromator and analyzer at the Brookhaven National Laboratory High Flux Beam Reactor. Most of the data were taken with a fixed final energy of 14.7 meV and 40 minute collimations throughout, yielding an energy resolution (full-width-at-half-maximum) of 1 meV. Higher harmonic contamination of the scattered beam was reduced by a PG filter after the sample. The polarized beam measurements were made on a modified triple axis spectrometer using procedures described elsewhere[5,6]. The inelastic spectrum was measured at constant Q with a fixed final energy of 30.5 meV and collimations of 40'-80'80'-open, which yielded an energy resolution (FWHM) of 5 meV. All measurements described here were made with the momentum transfer, Q long c*, so that we probe the generalized susceptibility in the ab plane.

The fundamental result of this work is shown in Fig. 1. Here we plot the scattered intensity measured with unpolarized neutrons at a fixed energy loss of 8 meV as a function of momentum transfer $Q = (0,0,Q_Z)$ at 1.2K. Q_Z is expressed in reciprocal lattice units with $c^* = 2\pi/c = 1.28$ Å^{-1}. The sharp maxima at $Q_Z = 1.76$ and 2.24 correspond to a pair of longitudinal acoustic phonons expected at this wavevector and energy transfer. The underlying diffuse scattering clearly peaks at $Q_Z = 1$ and $Q_Z = 3$ and has a minimum at $Q_Z = 2$. The fact that the maximum at $Q_Z = 1$ is larger than that at $Q_Z = 3$ confirms the magnetic nature of the diffuse scattering. In fact, once corrections are made for differences in the illuminated volume of the sample at these two wavevectors, the intensity difference is in good agreement with the decrease expected for the U 5f magnetic form factor.[7] The fact that the observed scattering has minima at $Q_Z = 2n$ and maxima at $Q_Z = 2n + 1$, indicates the presence of AF correlations between the 2 U ions in the 2 (c) positions of the unit cell (space group $P6_3/mmc$).

The substantial difference between the magnetic scattering at $Q_Z = 1$ and $Q_Z = 2$, and the temperature dependence of this difference is best shown in Figure 2. Here we display a series of energy scans obtained at these two wavevectors using unpolarized neutrons. At 30K (Fig. 2c) there is no noticeable difference between the magnetic scattering at $Q_Z = 1$ and $Q_Z = 2$. However, as the temperature is reduced below T_χ (Fig. 2b) a noticeable difference which increases with decreasing temperature (Fig. 2a), is observed. The temperature of the onset of these AF spin fluctuations correlates well with T_χ. Therefore we conclude that the decrease measured in the bulk susceptibility below 18K is due to the onset of AF spin fluctuations.

The paramagnetic scattering spectra shown in Fig. 2 may be described by a cross-section per magnetic ion:

$$\frac{d^2\sigma}{d\Omega d\omega} = \gamma_o^2 \frac{k_f}{k_i} \left|f(\underset{\sim}{Q})\right|^2 (1-e^{-h\omega/kT})^{-1} \frac{\chi''(\underset{\sim}{q},\omega)}{\pi} \tag{1}$$

where the symbols have their usual meanings[8]. Aeppli et al.[9,4] have shown that the energy dependence of the response in UPt_3 is well characterized by:

$$\chi''(\underset{\sim}{q},\omega) = \chi'(\underset{\sim}{q}) \frac{\Gamma(\underset{\sim}{q})}{\Gamma(\underset{\sim}{q}) + i\omega(\underset{\sim}{q})} \tag{2}$$

where χ ($\underset{\sim}{q}$) is the real, zero-frequency $\underset{\sim}{q}$-dependent susceptibility and $\Gamma(\underset{\sim}{q})$ is the half-width at half-maximum of the quasielastic response at that wavevector. Therefore, $\Gamma(\underset{\sim}{q})$ is a measure of the characteristic energy scale for the spin fluctuations at wavevector $\underset{\sim}{q}$. The reader will notice that the spectra of Fig. 2 apparently peak at finite energy transfer; this follows because for $kT << \Gamma(\underset{\sim}{q})$ the scattering described by Eqns. (1) and (2) peaks at $\omega_o = \Gamma(\underset{\sim}{q})$. As the temperature increases, $\omega_o \to 0$, hence the maximum at 30K (fig. 2c) appears at somewhat smaller energy transfer.

The data in Fig. 2 were fit using Eqn. (2) corrected for the instrumental resolution and the higher harmonic content of the incident beam. These fits are shown as the solid and dashed lines in this figure. At 4.2K the fit to the scattering at $Q_Z = 1.05$ yielded a quasielastic half-width of 4.3 ± 0.3 meV (~50K). Fits to the data at $Q_Z = 2$ yield approximately the same quasielastic half-width with somewhat larger uncertainty since there is no distinct maximum observed in these inelastic spectra.

Figure 3 displays the magnetic scattering intensity obtained at $Q_Z = 1.1$ in the polarized beam measurement.[5] The difference in the scattering measured with the neutron polarization along the scattering vector (HF) and perpendicular to the scattering plane (VF) yields one-half of the magnetic cross-section measured with unpolarized neutrons. However, this method unambiguously isolates the magnetic contribution from the nuclear background. A fit to these data using Eqn. (2) yields a quasielastic half-width of 6.4 ± 2.2meV, consistent with the unpolarized beam value. The inset of Fig. 3 shows the polarized beam measurement on polycrystalline UPt_3 by Aeppli et al.[9] They reported an energy width of 10 ± 2meV, somewhat larger than, although not far from, the value obtained here.

DISCUSSION

The observation of AF spin fluctuations in the low temperature normal state of UPt_3 is inconsistent with the analogy often made between this HFS and liquid ^3He, and may have important consequences for the description of the pairing mechanism responsible for the superconductivity in this system. Recent work by several theorists[10] has indicated that AF spin fluctuations tend to suppress odd parity superconductivity. We are presently collecting neutron scattering data with $\underset{\sim}{Q}$ along a* or b* to observe the character of the spin fluctuations with wavevectors in the basal planes.

Previous magnetic scattering measurements[9] on a polycrystalline sample of UPt_3 yielded information about the energy scale of the spin fluctuations in this system. However, information concerning the $\underset{\sim}{q}$-dependence of $\chi(\underset{\sim}{q},\omega)$ is lost in the powder average. The larger quasielastic half-width reported in that work may result from a somewhat greater energy scale for the spin fluctuations along a* or b*, since only the average value is measured in a powder. A previous single crystal neutron scattering measurement which indicated the existence of antiferromagnetic correlations was subsequently reported by the authors to be erroneous.[11] However, Aeppli et al.[12] have recently observed AF correlations in another HFS, $CeCu_6$. Cox et al.[13] have described the antiferromagnetic structure of U_2Zn_{17}. Perhaps, at least for the systems listed above, the outstanding features of the HFS are in part due to proximity to an antiferromagnetic instability.

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Figure Captions

1. Q-dependence of the paramagnetic scattering taken at a constant energy transfer of 8 meV. Closed triangles show the background level measured on the energy gain side. The two sharp peaks correspond to phonons, and the lines are included as a guide to the eye. (after Ref. 4)
2. Spectra taken at constant $Q_Z = 1.05$ and 2, after background subtraction at a) 4.2K, b) 10K and c) 30K. The lines represent the best fit to the data as described in the text. (after Ref. 4)
3. Inelastic magnetic scattering measurement at $Q_Z = 1.1$ using a polarized beam (after Ref. 5). The inset shows a previous measurement on a polycrystalline sample by Aeppli et al.[9].

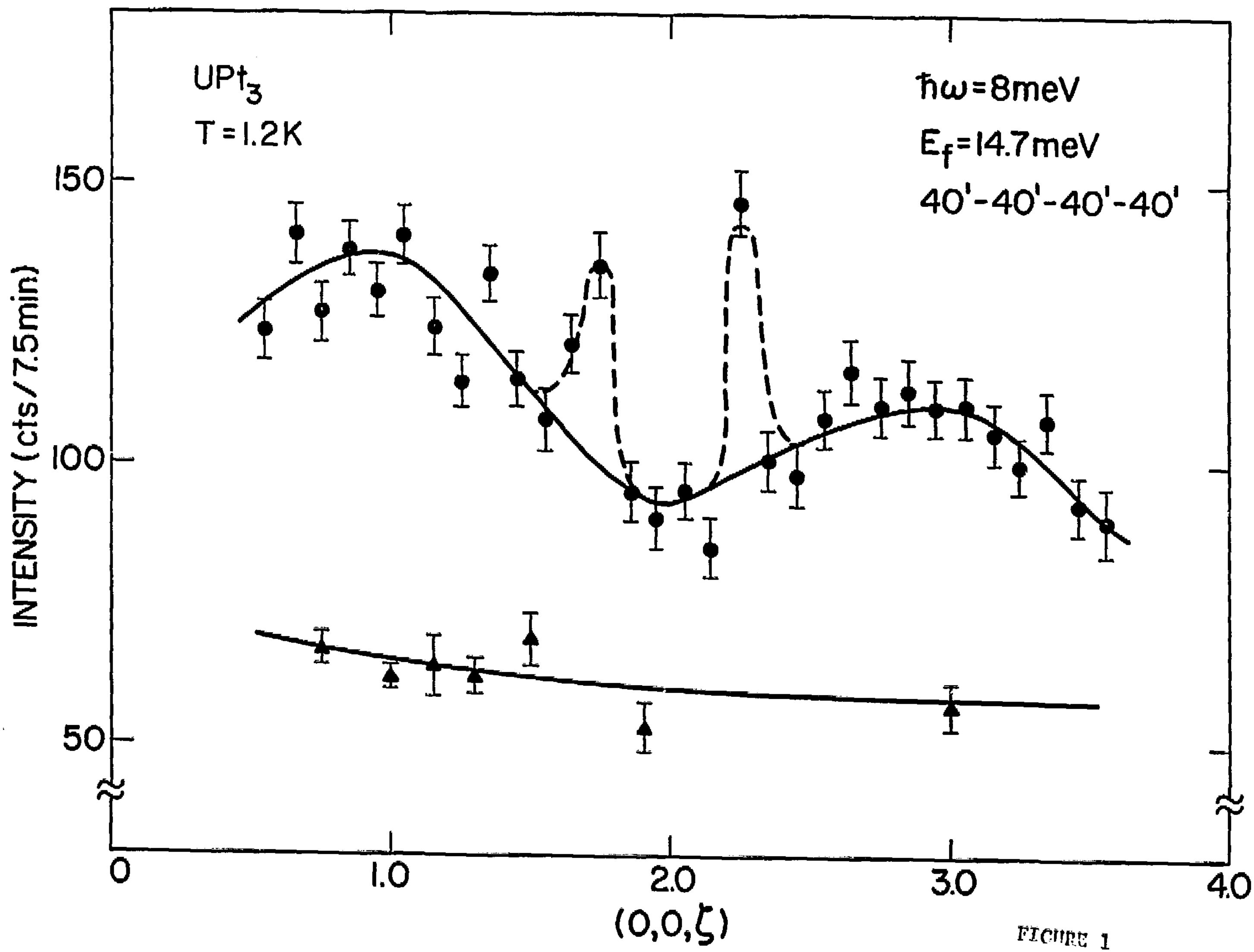
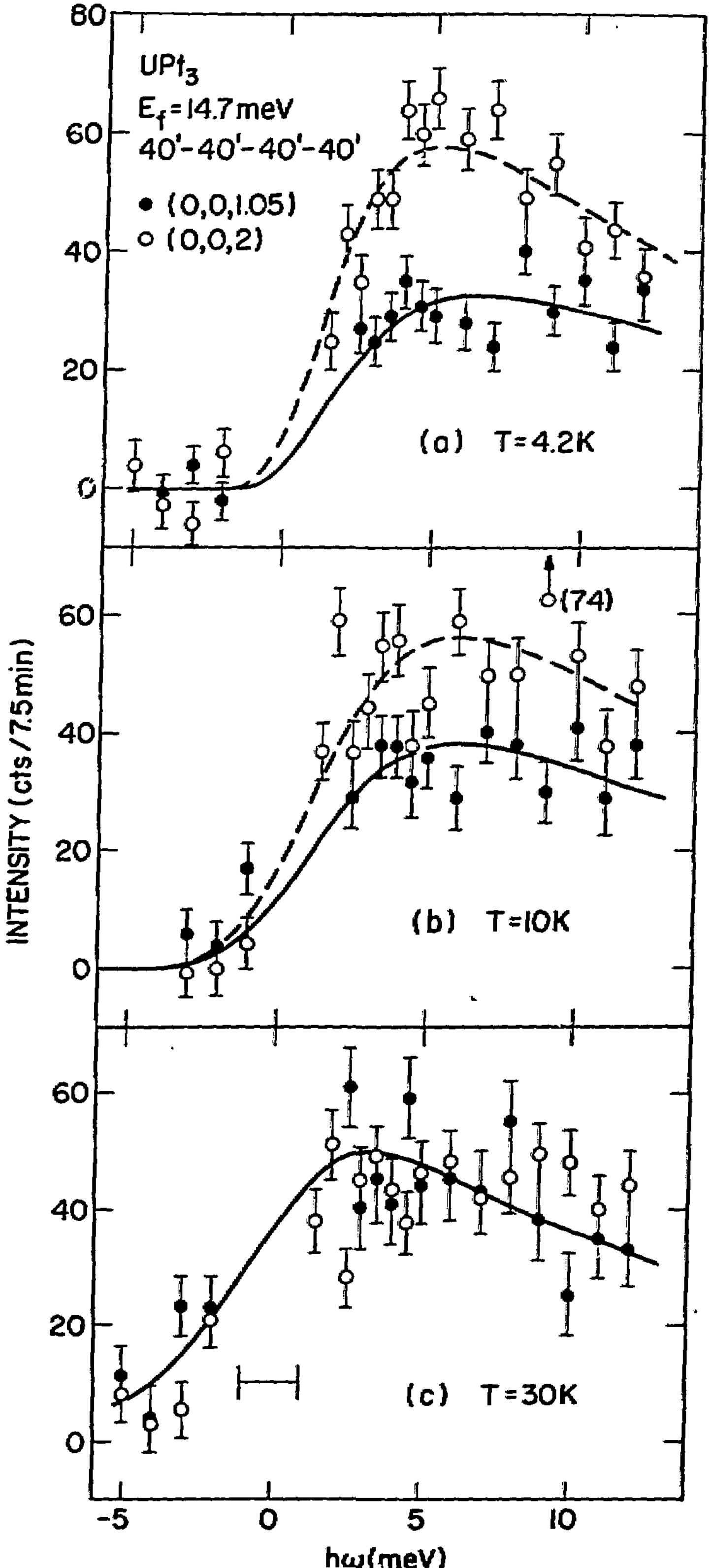
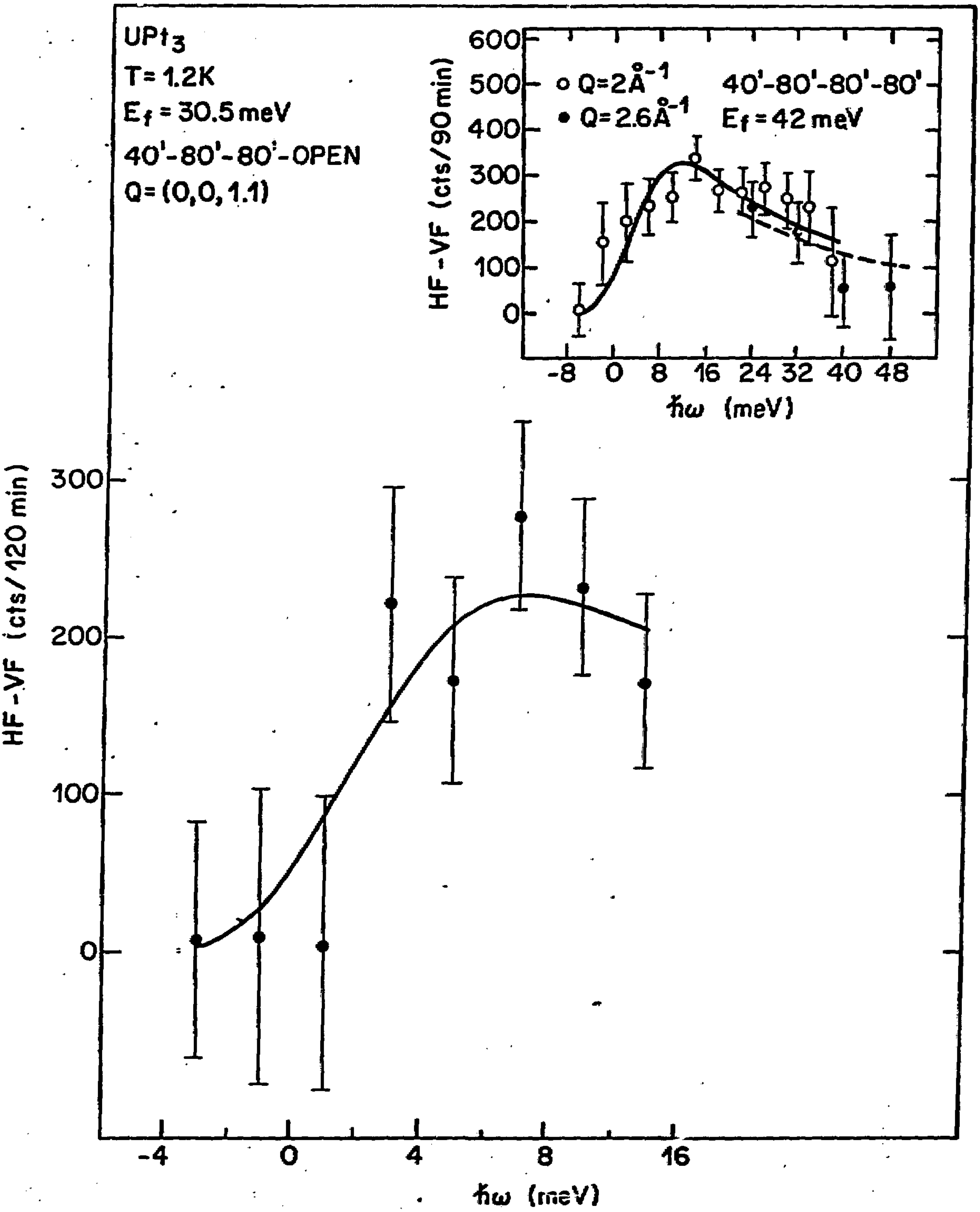

UPt3
T = 1.2K
ħω=8meV
Ef=14.7meV
40'-40'-40'-40'
INTENSITY (cts/7.5min)
150
100
50
0
1.0
2.0
3.0
4.0
(0,0,ζ)

FIGURE 1



UPt3
Ef=14.7meV
40'-40'-40'-40'
(0,0,1.05)
(0,0,2)
(a) T=4.2K
(74)
(b) T=10K
(c) T=30K
INTENSITY (cts/7.5min)
hω(meV)

FIGURE 2



UPt3
T= 1.2K
Ef = 30.5 meV
40'-80'-80'-OPEN
Q= (0,0,1.1)
HF - VF (cts/120 min)
ħω (meV)
HF - VF (cts/90 min)
o Q=2Å-1
• Q=2.6Å-1
40'-80'-80'-80'
Ef = 42 meV
ħω (meV)

FIGURE 3